

ACCGE-18 ABSTRACT

Existence and Stability of Menisci in Detached Bridgman Growth

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Abstract

Detached growth, also referred to as dewetted growth, is a Bridgman crystal growth process in which the melt is in contact with the crucible wall but the crystal is not. A meniscus bridges the gap between the top of the crystal and the crucible wall. The Young-Laplace capillary equation was used to calculate the crystal radii of detached states as a function of the pressure differential across the meniscus. The detached states depend on the contact angle of the melt with the crucible wall, the growth angle of the melt with respect to the solidifying crystal, and the Bond number. A static stability analysis was performed on the calculated detached states. The stability criterion was the sign of the second variation of the potential energy upon admissible meniscus shape perturbations. The conditions considered corresponded to the growth of Ge and InSb, in both terrestrial and microgravity conditions. Stability was found to depend significantly on whether the interior surface was considered to be microscopically rough or smooth, corresponding to pinned or unpinned states. It was also found that all meniscus shapes which are single-valued functions of the radius are statically stable in a microgravity environment.



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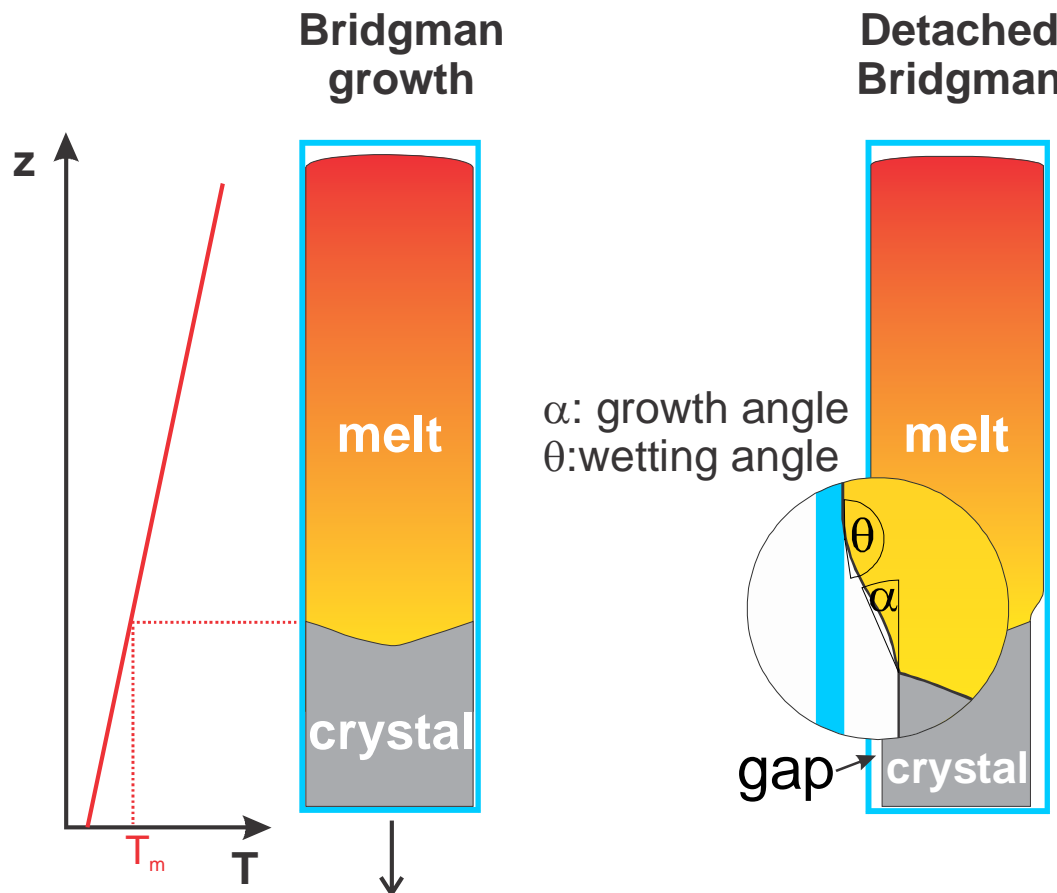
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Principles of Detached Bridgman Growth

Sufficient condition for detachment^{1,2}:
 $(\alpha + \theta \geq 180^\circ)$



Advantages

- No sticking of the crystal to the ampoule wall
- Reduced stress
- Reduced dislocations
- No heterogeneous nucleation by the ampoule
- Reduced contamination

¹V. S. Zemskov:
Fiz. Khim. Obrab. Mater. 17 (1983) 56

²T. Duffar, I. Paret-Harter, P. Dusserre:
J. Crystal Growth 100 (1990) 171.



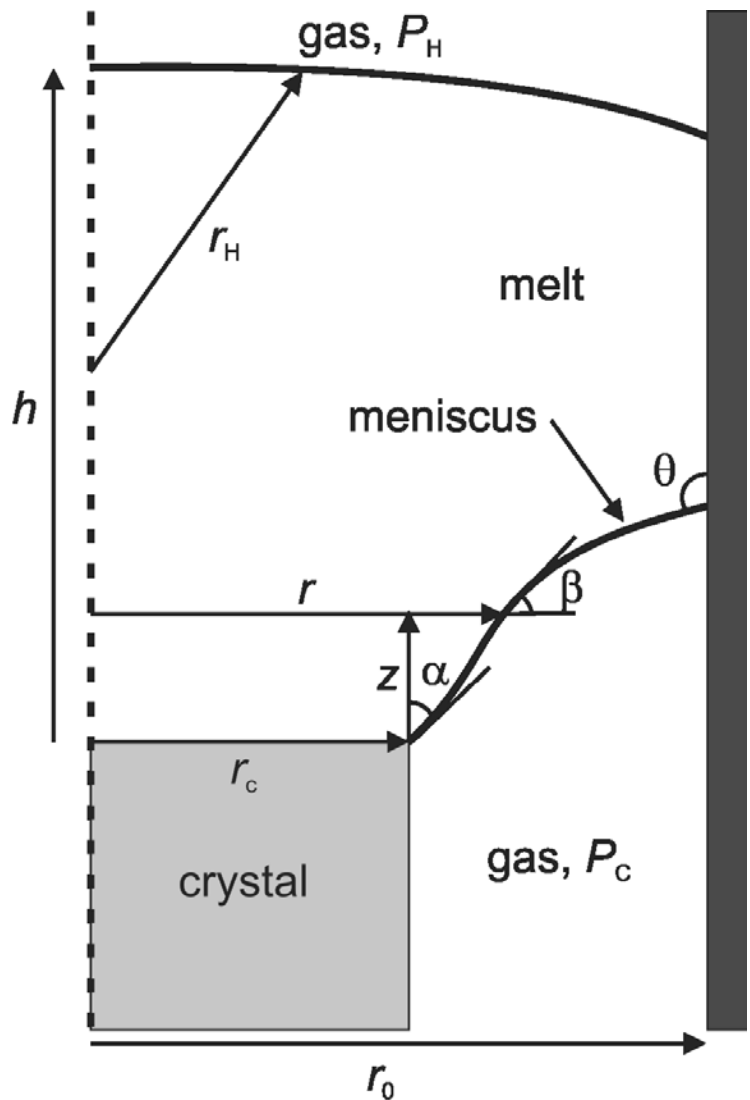
Motivating Questions



- What are the conditions to achieve detached growth?
- What are the necessary conditions to establish a meniscus between the crystal and ampoule wall?
- How does the existence of detached growth depend on the pressure differential across the meniscus, the growth angle, the contact angle and the Bond number?
- What are the conditions for the static stability of a meniscus?
- What are the conditions for the dynamic stability of a meniscus?



Schematic Diagram of Detached Solidification



M. P. Volz, K. Mazuruk, *Journal of Crystal Growth* 321 (2011) 29-35



Calculation of Meniscus Shapes



$$\frac{\frac{d^2 z}{dr^2}}{\left(1 + \left(\frac{dz}{dr}\right)^2\right)^{3/2}} + \frac{\frac{dz}{dr}}{r \left(1 + \left(\frac{dz}{dr}\right)^2\right)^{1/2}} = \Delta P - Bz(r)$$

Young-Laplace Equation

$$\Delta P = \frac{\Delta P_m r_0}{\sigma}, \quad \Delta P_m = P_H - P_C + \rho g h + 2 \frac{\sigma}{r_H}$$

ΔP : Dimensionless pressure differential across the meniscus

$$B = \frac{\rho g_0 r_0^2}{\sigma} \quad \begin{array}{l} B = 3.248; \text{ Ge, } r_0 = 6 \text{ mm} \\ B = 4.651; \text{ InSb, } r_0 = 5.5 \text{ mm} \end{array}$$

B : Bond number; ratio of gravity force to surface tension force

$$\frac{\partial r}{\partial s} = \cos \beta, \quad \frac{\partial z}{\partial s} = \sin \beta, \quad \frac{\partial \beta}{\partial s} = -\frac{\sin \beta}{r} + \Delta P - Bz$$

Set of 3 coupled differential equations

Boundary Conditions

$$z(0) = 0; \quad \beta(0) = 90^\circ - \alpha;$$

$$\beta(1) = \theta - 90^\circ; \quad r(1) = 1$$

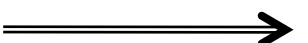
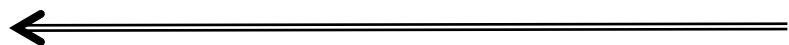
α : growth angle

θ : contact or wetting angle

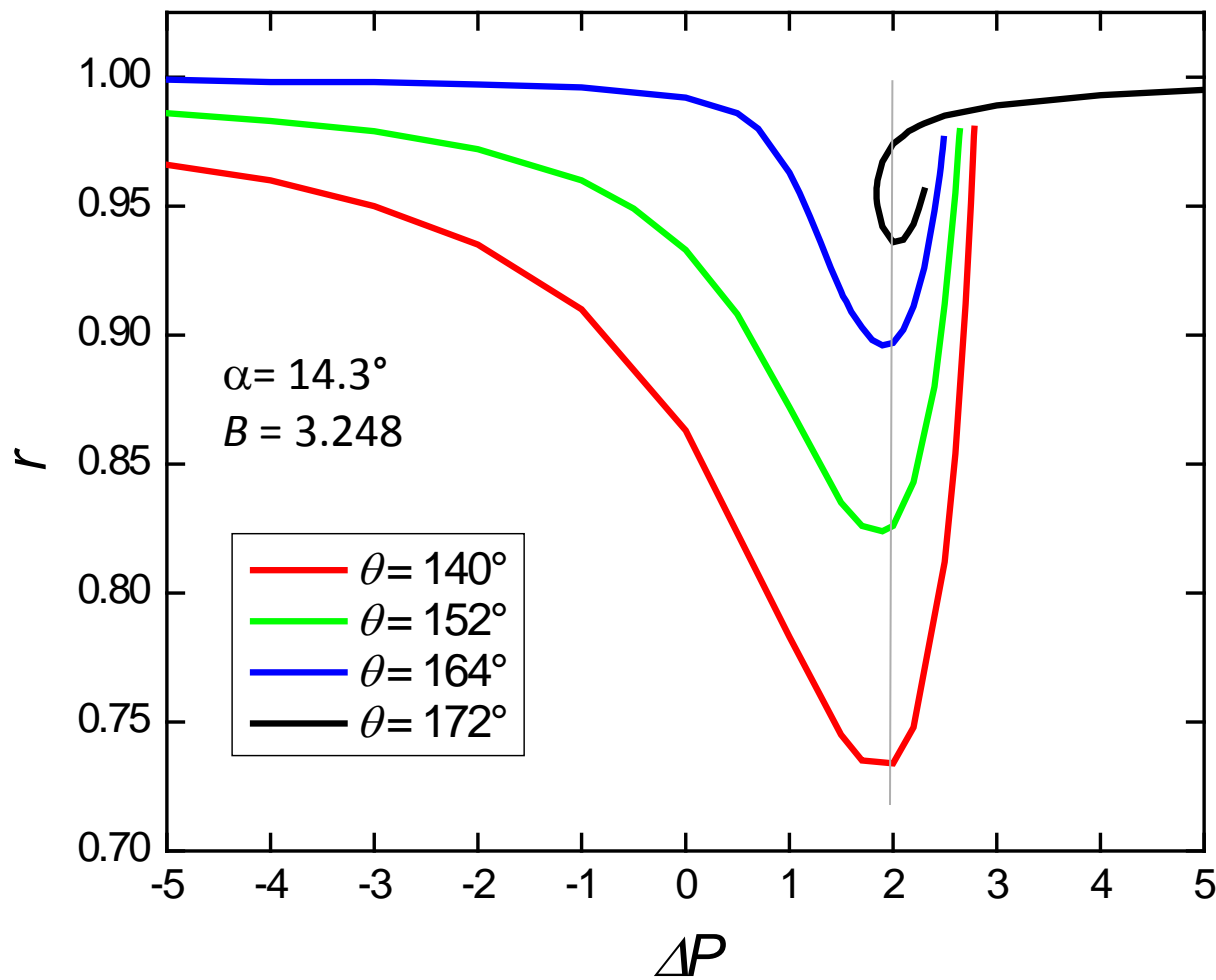


Gap Width vs. Pressure Differential (Ge at 1g)

$\theta + \alpha < 180^\circ$



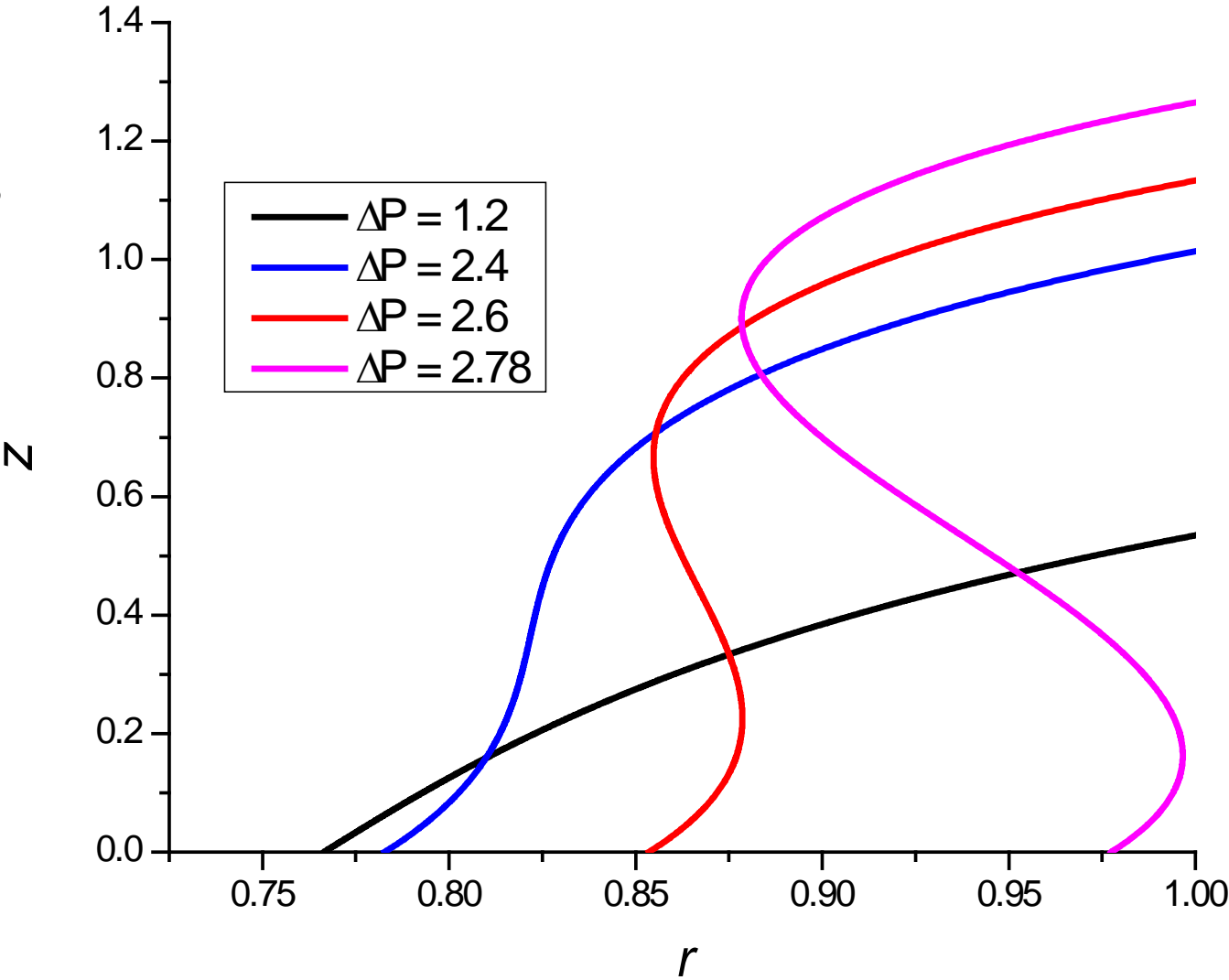
$\theta + \alpha > 180^\circ$





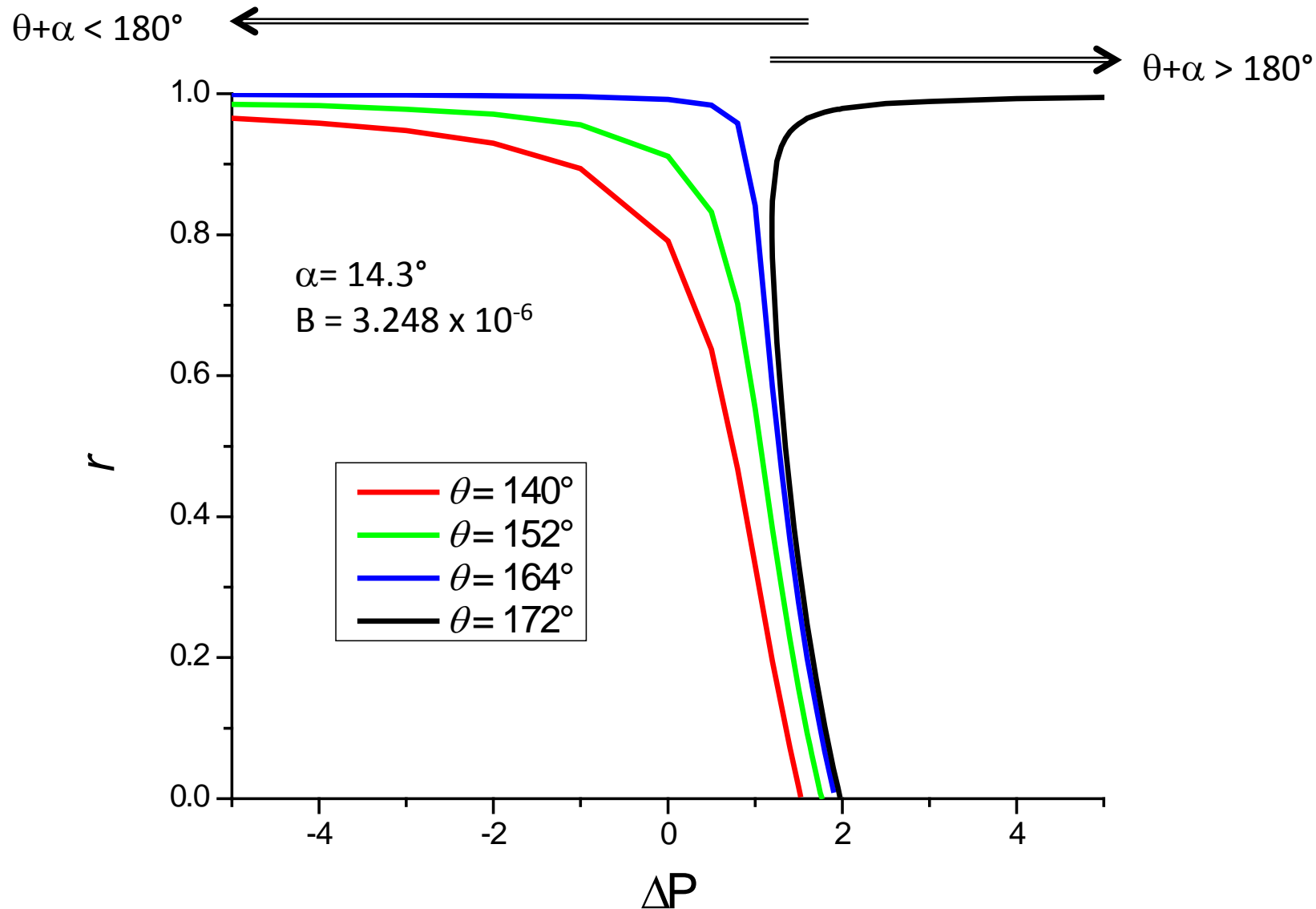
Meniscus Shapes vs. ΔP for $\theta = 140$

$\alpha = 14.3^\circ$
 $B = 3.248$





Gap Width vs. Pressure Differential (Ge at $g = 1 \times 10^{-6} g_0$)



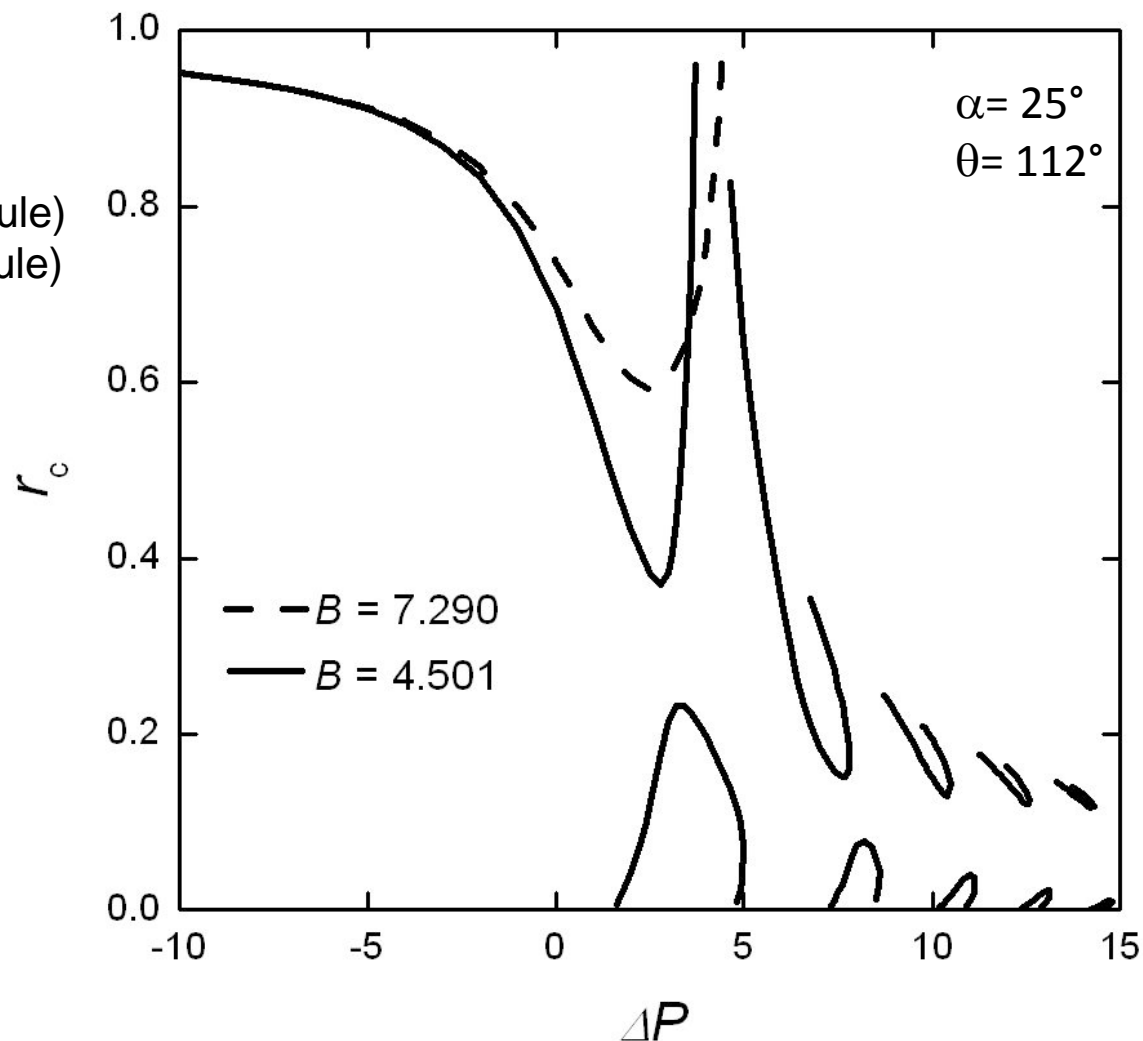


Gap Width vs. Pressure Differential (InSb at 1g)

InSb at 1g

$B = 7.209$ (14 mm diam. ampoule)

$B = 4.501$ (11 mm diam. ampoule)





Static Stability Analysis

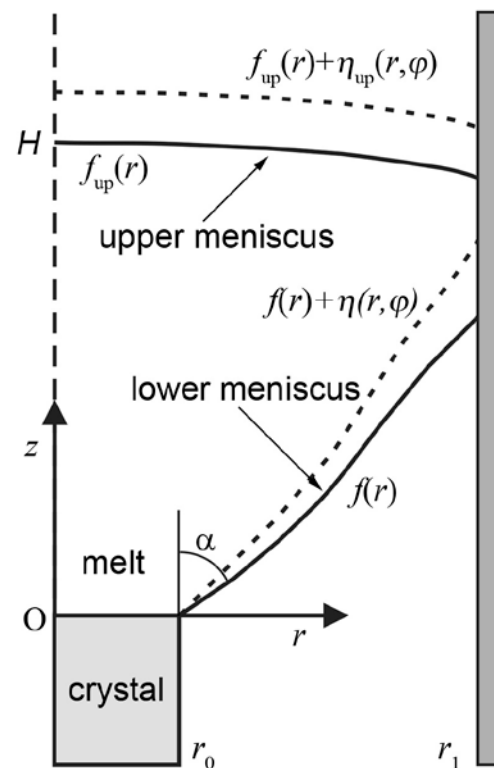


$$\Delta U = -P_H \Delta V_H - P_C \Delta V_C + \sigma \Delta S_H + \sigma \Delta S_C + \sigma_{gc} \Delta \Sigma_{gc} + \sigma_{mc} \Delta \Sigma_{mc} + g \rho \Delta \left(\int z dV \right)$$

ΔU potential energy change
 P_H, P_C upper and lower gas pressures
 $\Delta V_H, \Delta V_C$ upper and lower gas volumes
 $\Delta S_H, \Delta S_C$ upper and lower meniscus surfaces
 σ melt surface tension
 σ_{gc}, σ_{mc} gas-crucible and melt-crucible interface energy
 g gravitational acceleration
 ρ, V melt density and volume

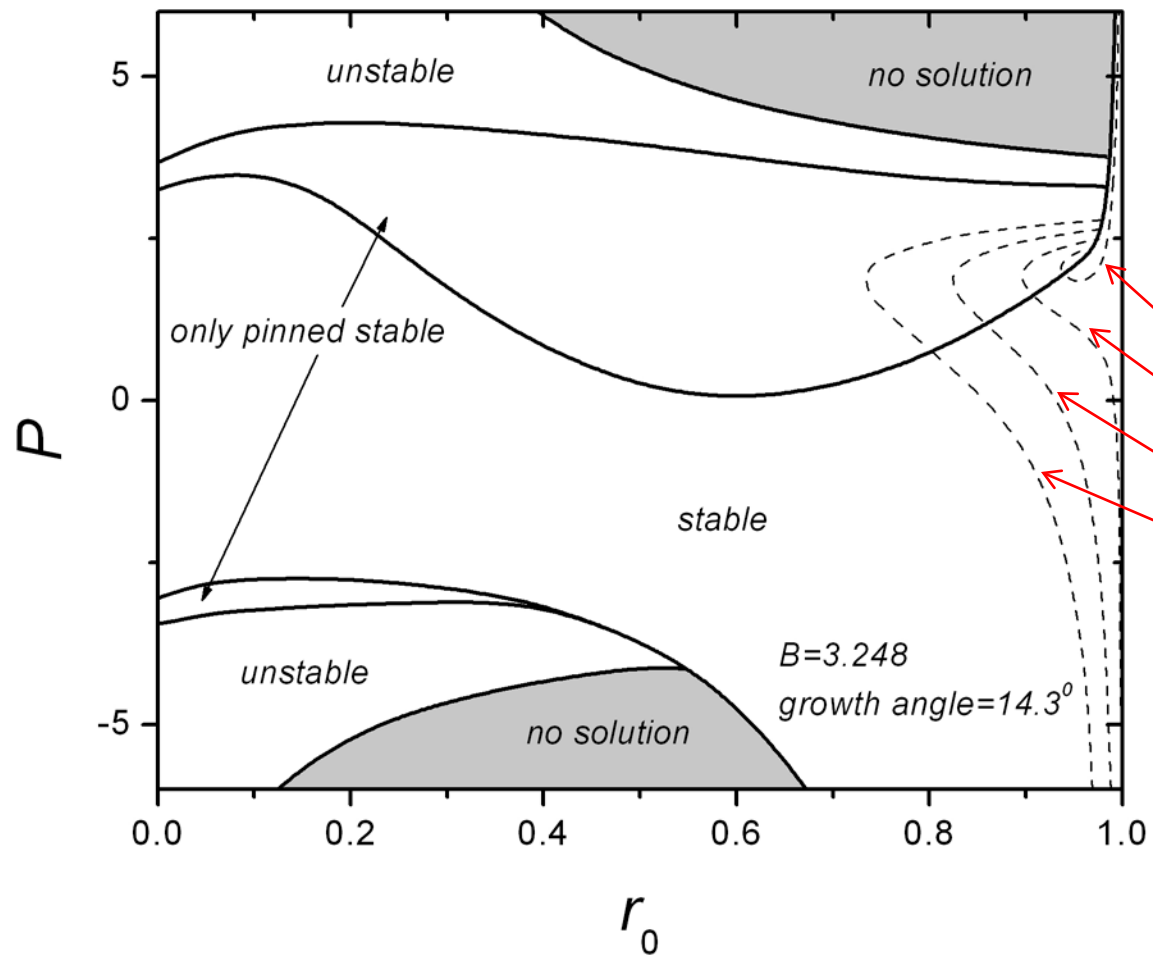
Analysis Approach

- Minimize the potential energy
- Consider both pinned and non-pinned menisci at the crucible wall
- Consider both axisymmetric and non-axisymmetric perturbations





Stability Map for Ge at 1g: Axisymmetric modes

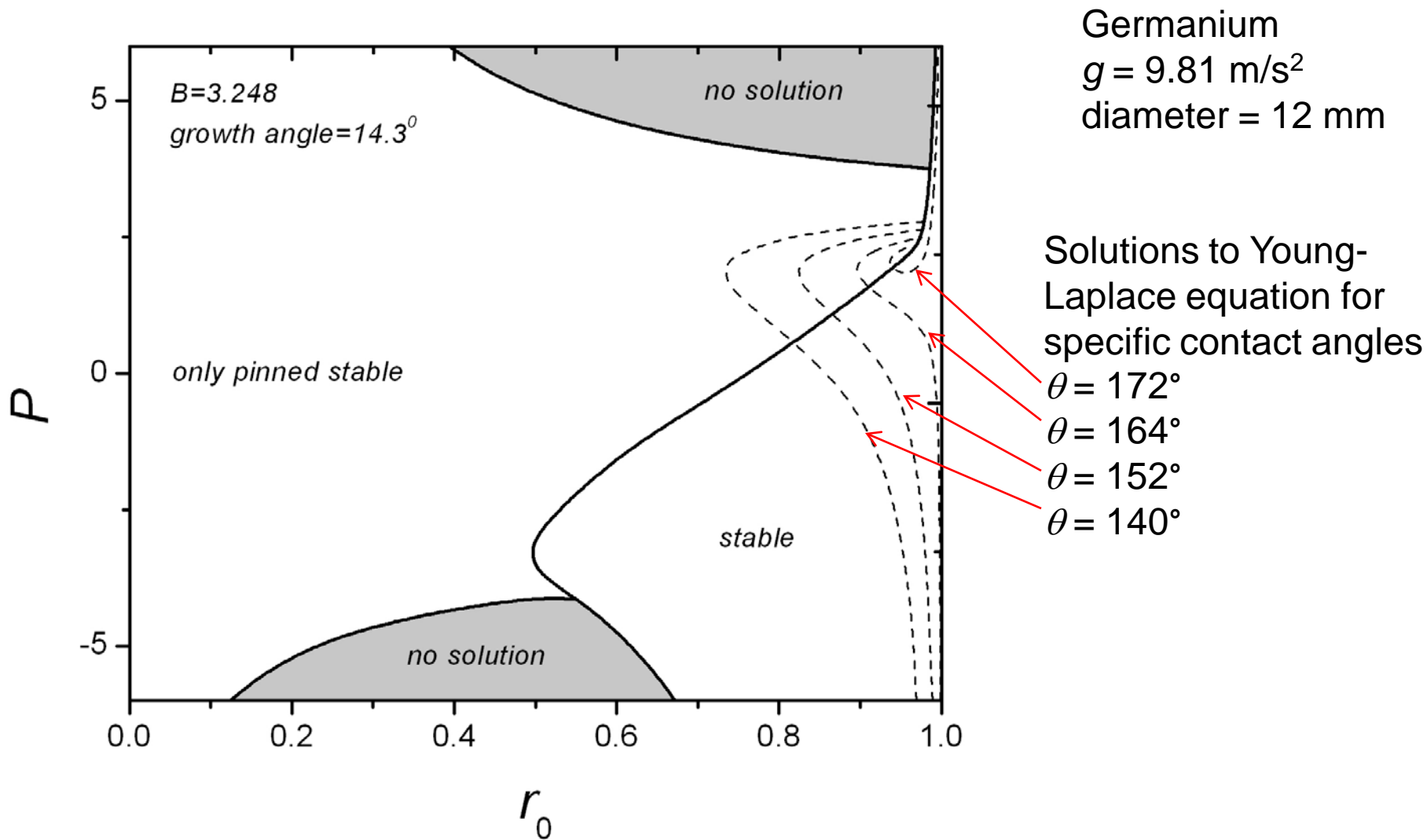


Germanium
 $g = 9.81 \text{ m/s}^2$
diameter = 12 mm

Solutions to Young-Laplace equation for specific contact angles

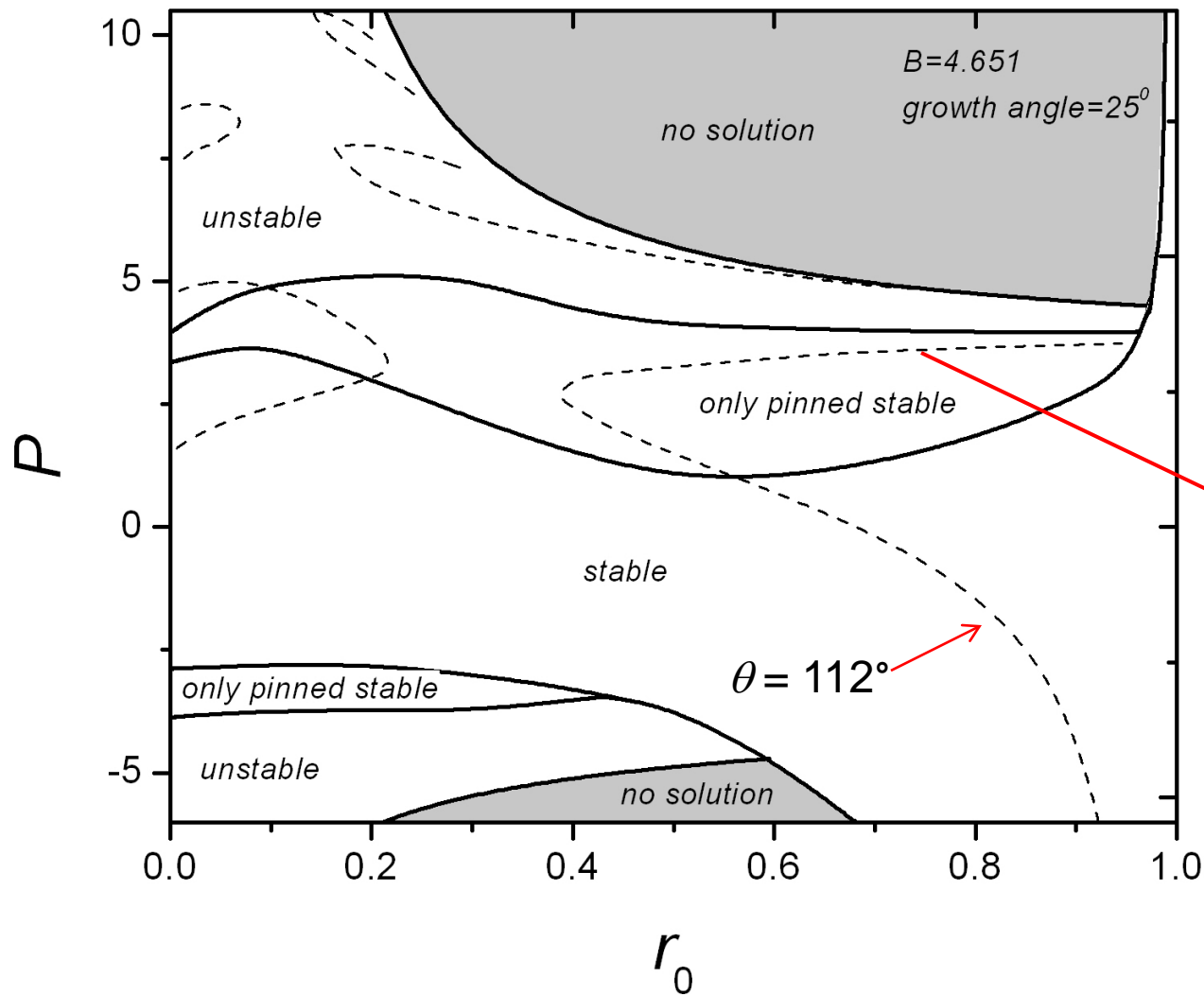


Stability Map for Ge at 1g: Non-axisymmetric Modes

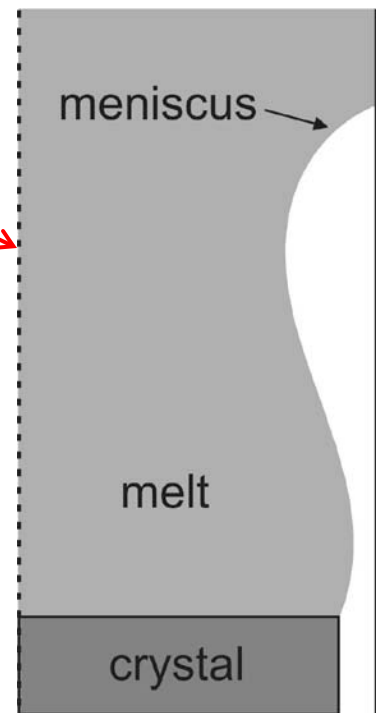




Stability Map for InSb at 1g: Axisymmetric Modes

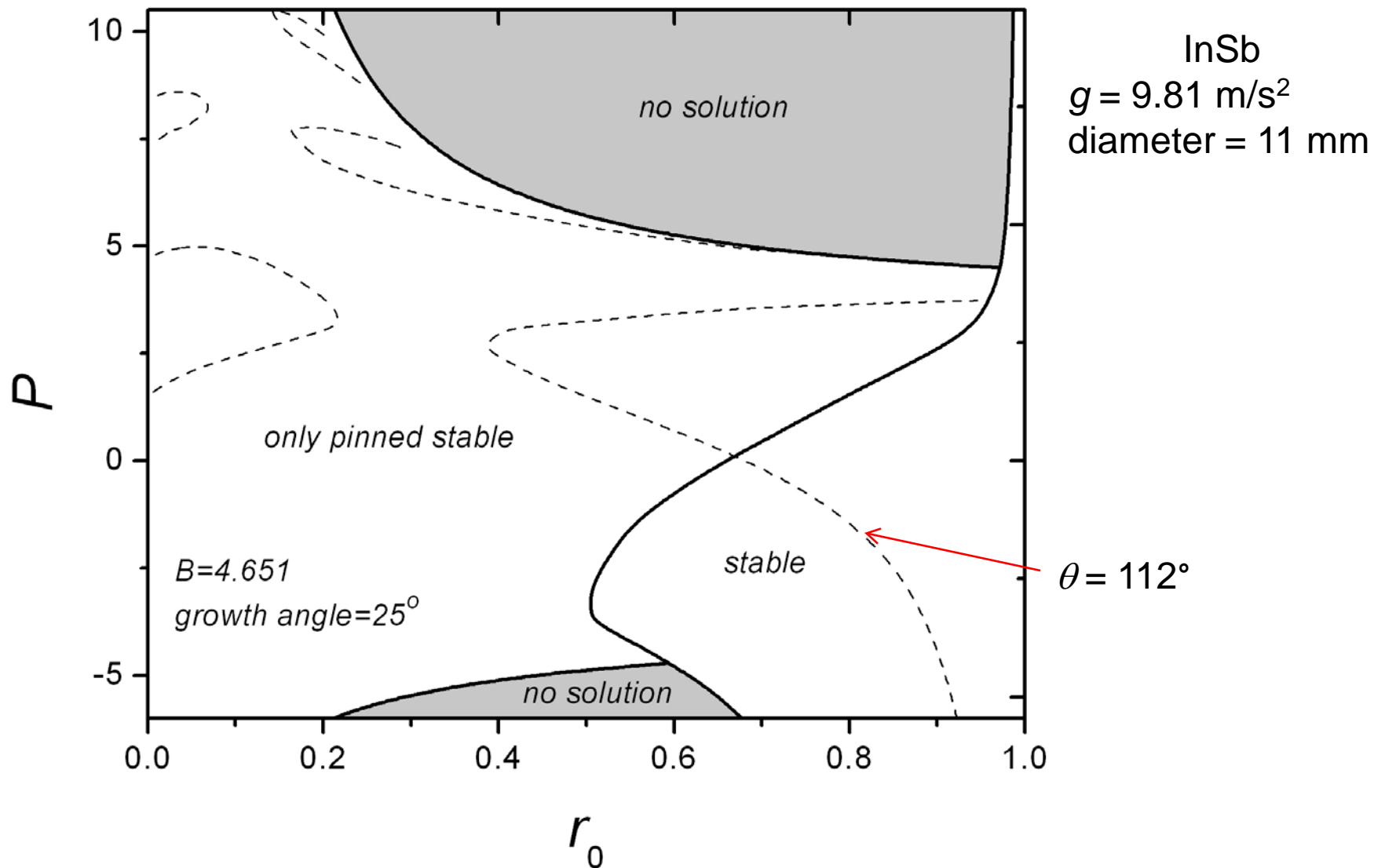


InSb
 $g = 9.81 \text{ m/s}^2$
diameter = 11 mm





Stability Map for InSb at 1g: Non-axisymmetric Modes





Conclusions



- Detached growth requires a meniscus. The existence and shape of menisci depend on the growth and contact angle, the pressure differential, and the Bond number.
- Whether $\theta + \alpha$ is less than or greater than 180° is the determining factor in whether menisci exist at large positive or negative pressure differentials.
- All menisci in microgravity are stable.
- Menisci can have several stability classifications: always unstable, only stable when pinned, always stable.
- Non-axisymmetric perturbation modes are more dangerous than axisymmetric perturbation modes.
- Calculations indicate that stable detached growth can be achieved in terrestrial conditions but that accurate knowledge and control of the pressure differential across the meniscus is required.